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Optimization of the Nonradiative Lifetime of Molecular-Beam-Epitaxy (MBE)-Grown Undoped GaAs/AlGaAs Double Heterostructures (DH)

by P. Folkes, H. Hier, B. VanMil, B. C. Connelly, and W. Beck

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14. ABSTRACT
In this report, we present results of an ongoing study aimed at measuring and optimizing the nonradiative lifetime and the internal radiative quantum efficiency of molecular beam epitaxy (MBE)-grown gallium arsenide (GaAs) solar cells that are grown at the U.S. Army Research Laboratory (ARL) using different substrate growth temperatures and arsenic (As)/gallium (Ga) flux ratios to determine the growth parameters that maximize the bulk GaAs minority carrier nonradiative lifetime. We report a significant increase in the nonradiative lifetime and the internal radiative quantum efficiency of MBE-grown GaAs/aluminum gallium arsenide (AlGaAs) double heterostructure (DH) structures grown at ARL with a growth temperature of 595 °C and an As/Ga flux ratio = 15. Our results show that the nonradiative lifetime and internal radiative quantum efficiency of the DH structures grown using these parameters are comparable to those of the highest quality reported MBE-grown GaAs.

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1. Introduction

The theoretical prediction for the maximum energy-conversion efficiency in photovoltaic solar cells assumes that energy loss is dominated by radiative recombination of minority carriers (*1*). This means that a semiconductor solar cell structure with internal radiative quantum efficiency near unity is required to achieve the predicted maximum energy-conversion efficiency. The internal radiative quantum efficiency $\eta = \tau_{nr}/(\tau_r + \tau_{nr})$, where τ_r is the minority carrier radiative recombination lifetime and τ_{nr} is the nonradiative recombination lifetime. Ideal radiative quantum efficiency requires that $\tau_{nr} >> \tau_r$. Recent measurements of the minority carrier lifetimes in p-gallium arsenide (GaAs) in GaAs/aluminum gallium arsenide (AlGaAs) double heterostructures (DHs) that were grown by molecular beam epitaxy (MBE) at the U.S. Army Research Laboratory (ARL) (*2*) show that $\eta = 0.68$. Published research results on GaAs/AlGaAs DHs that were grown at another laboratory (*3*) show that $\eta \geq 0.90$ for the highest-quality GaAs, indicating that the quality of GaAs grown by MBE at ARL can be significantly improved by increasing τ_{nr}. Published (*4*, *5*), research on the MBE growth and characterization of high-purity GaAs has shown that defect densities in GaAs grown by MBE at around 600 °C decrease as the arsenic (As)/gallium (Ga) flux ratio is decreased. However little has been published on the specific growth procedures that maximize the measured nonradiative lifetime of MBE-grown GaAs (*6*).

In this report, we present results of an ongoing study aimed at measuring the nonradiative lifetime of MBE-grown GaAs and optimizing the internal radiative quantum efficiency in the solar cells that are grown at ARL using different growth temperatures and As/Ga flux ratios to determine the growth parameters that maximize the bulk GaAs minority carrier nonradiative lifetime.

2. Experimental Technique and Results

Several GaAs/AlGaAs DHs with an undoped 20-µm GaAs layer were grown by MBE for studies of the nonradiative minority carrier lifetime. The structures were grown using different substrate growth temperatures and As/Ga flux ratios to determine the growth parameters that maximize the GaAs bulk nonradiative minority carrier lifetime. The basic heterostructure used to measure τ_{nr} is shown in figure 1. For times longer than around 40 ns, the photoluminescence (PL) decay of this DH structure is dominated by the τ_{nr} of the 20-µm undoped GaAs layer. Our approach to a high-efficiency solar cell uses a GaAs/AlGaAs DH structure with a roughly 2-µm GaAs active region on top of a Bragg reflector (BR) to take advantage of photon recycling effects. However, the effect of the growth of the BR on the deep level trap density of the GaAs active layer, which

determines the nonradiative lifetime, is unknown. A DH structure with a 20-µm GaAs layer on top of a distributed BR, comprising 10 periods of a 598-Å GaAs/726-Å $Al_{.8}Ga_{.2}As$ superlattice (figure 2), was also grown to study the effect of the Bragg reflector on the nonradiative lifetime of the 20-µm GaAs layer. The GaAs substrate temperature was calibrated using oxide desorption as observed by reflection high energy electron diffraction at 580 °C. As_4 was produced using an As cracker and the As flux was measured after ion gauge saturation. AlGaAs stoichiometry was calibrated using x-ray diffraction. Hall measurements were used to determine the carrier density and mobility of the 20-µm GaAs layer at 300 K and 77 K.

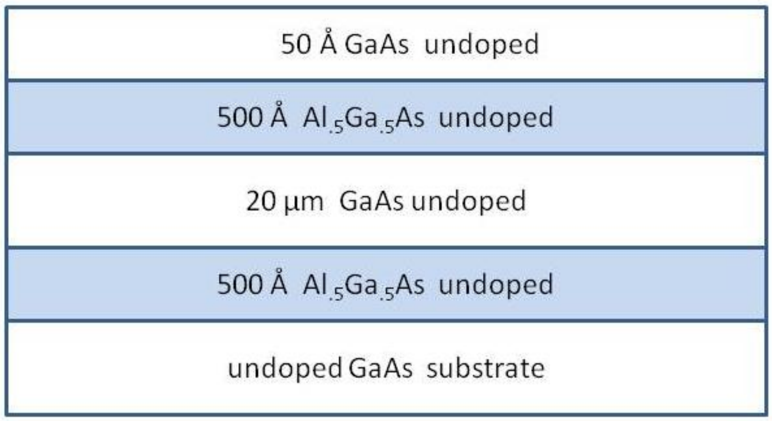

Figure 1. GaAs/AlGaAs DH.

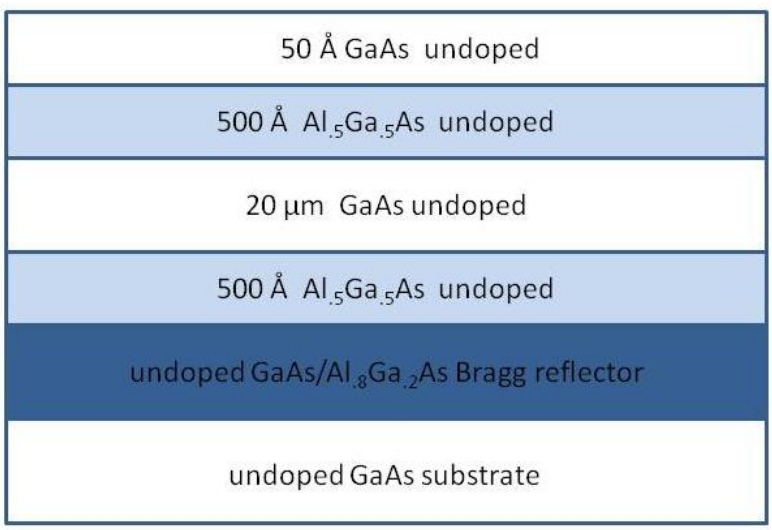

Figure 2. GaAs/AlGaAs DH structure with a BR.

Time-resolved photoluminescence (TRPL) measurements at 300 K were used to determine the nonradiative minority carrier lifetime τ_{nr}, of the GaAs/AlGaAs DH structures from the PL decay time, which is the effective minority carrier lifetime. Samples were excited using a 250-kHz repetition rate, ultrafast 632-nm laser (~1.5-mm beam diameter) that was derived from frequency doubling the output of a regenerative amplifier-pumped optical parametric amplifier. PL was

detected through a 700-nm long-pass filter, to minimize the laser scattering signal, with a fast 300-µm diameter silicon (Si) photodiode. Data were acquired on a PCI averager card. The system response was measured to be ~2 ns.

TRPL data and the various MBE growth parameters are shown in figures 3–7 and the mobility and carrier concentration determined from Hall measurements are shown in table 1.

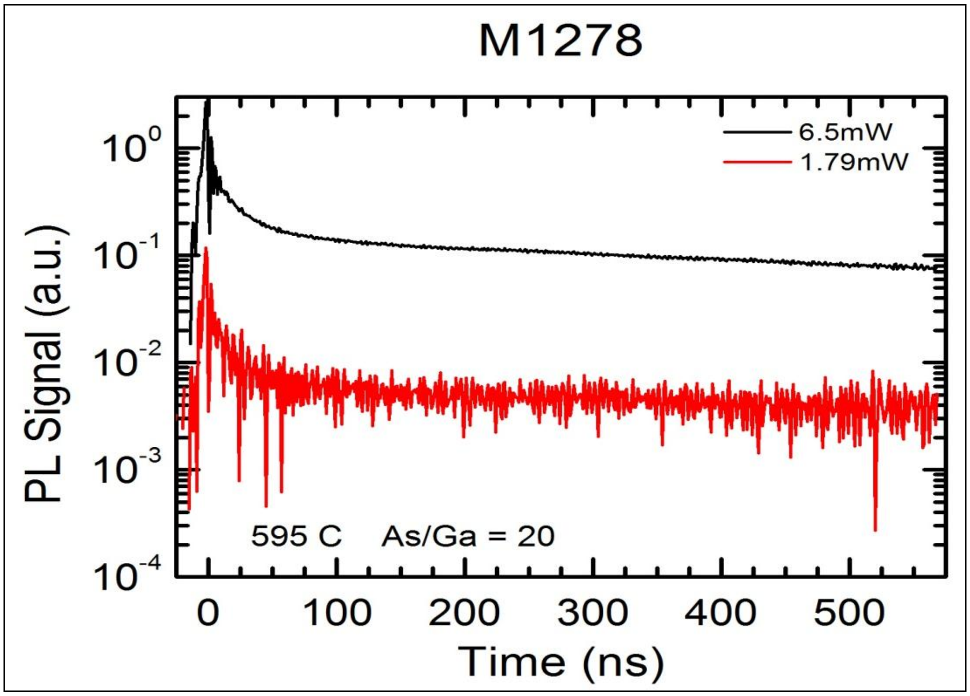

Figure 3. TRPL data for structure M1278 grown at 595 °C with an As/Ga ratio of 20.

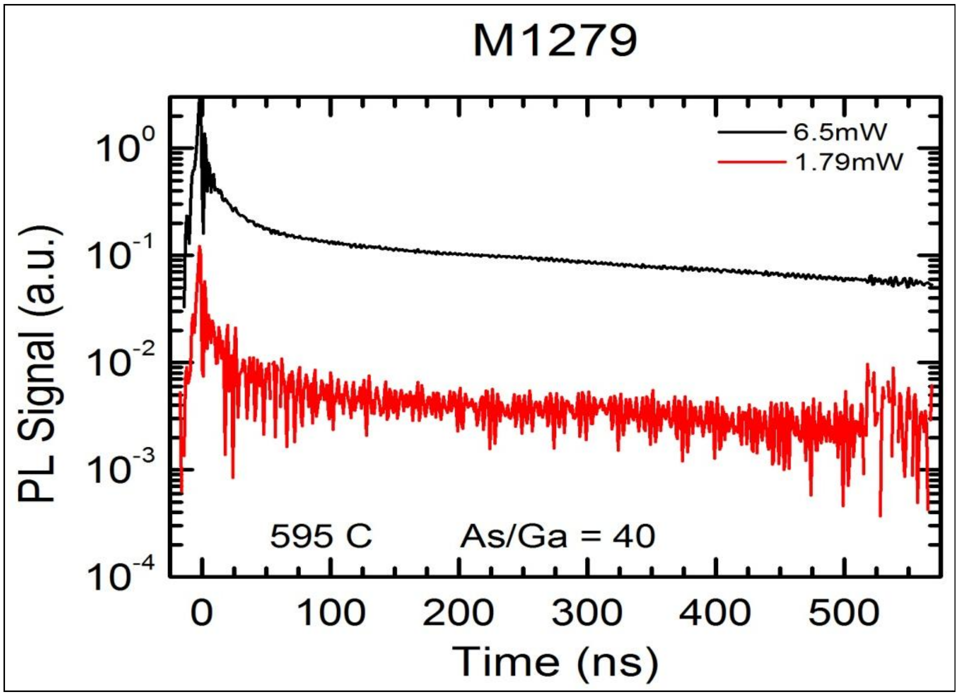

Figure 4. TRPL data for structure M1279 grown at 595 °C with an As/Ga ratio of 40.

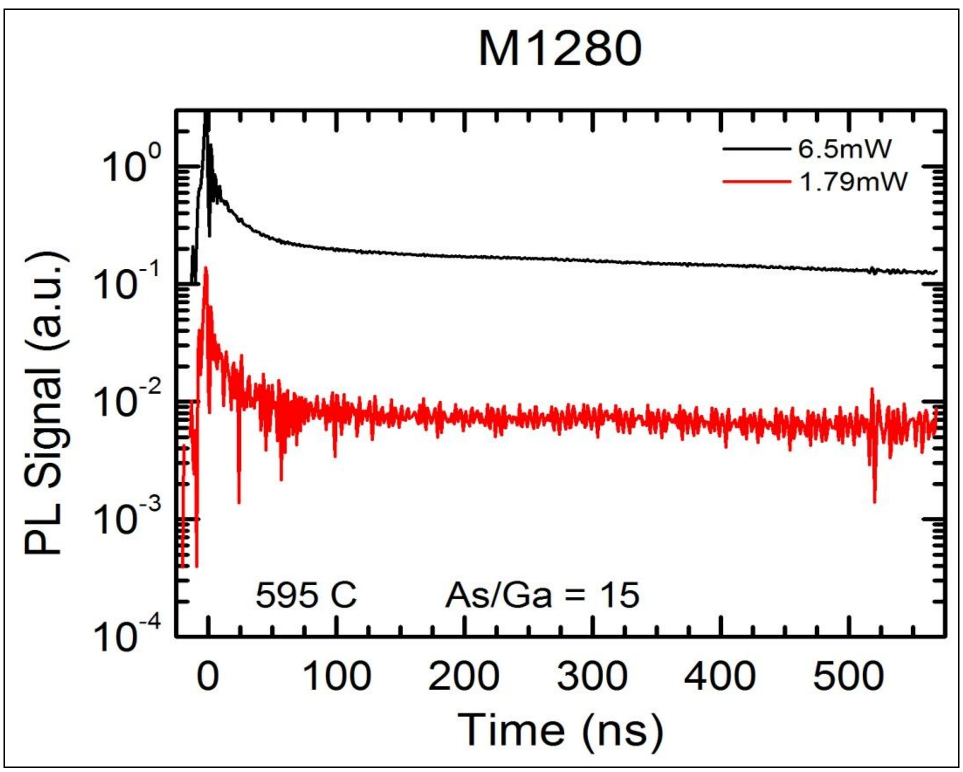

Figure 5. TRPL data for structure M1280 grown at 595 °C with an As/Ga ratio of 15.

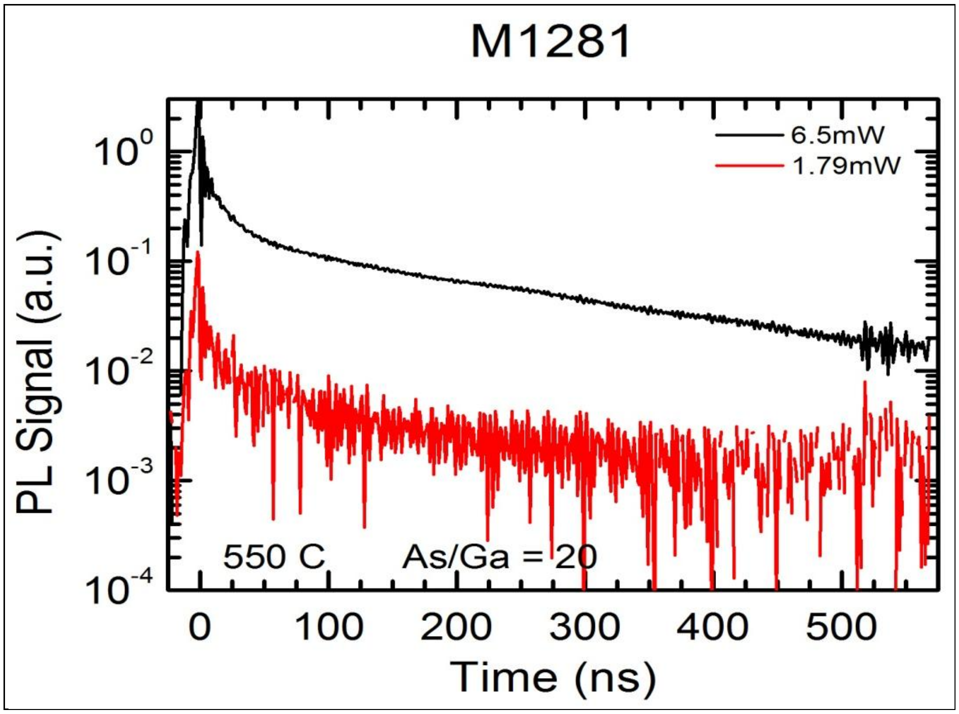

Figure 6. TRPL data for structure M1281 grown at 550 °C with an As/Ga ratio of 20.

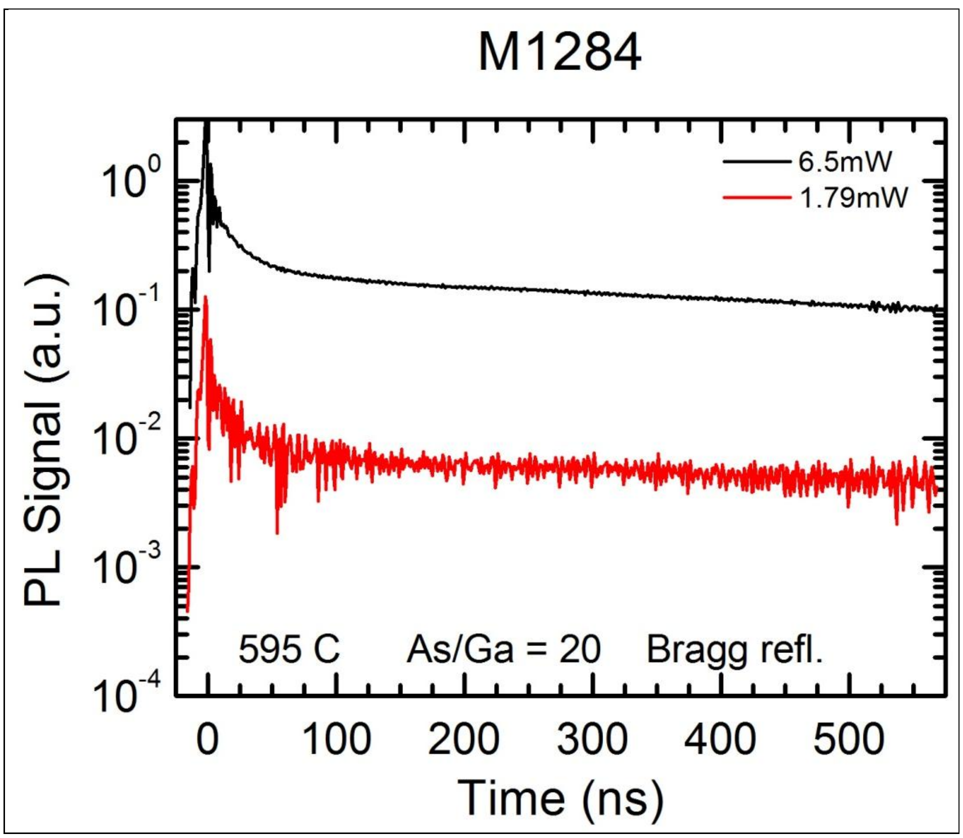

Figure 7. TRPL data for structure M1284 grown at 595 °C with an As/Ga ratio of 20 with a Bragg reflector.

Table 1. Hall measurement data.

20-µm GaAs w/ $Al_{.5}Ga_{.5}As$ Barriers

Sample #	Sheet Carrier Conc (cm^{-2})	Carrier Conc (cm^{-3})	Mobility (cm^2/ V-sec)	Temp (K)	As/Ga ratio
M1278	2.436 X 10^{11}	1.554 X 10^{14}	371	300	20
M1278	1.395 X 10^{11}	9.614 X 10^{13}	9046	77	20
M1279	8.430 X 10^{10}	6.364 X 10^{13}	316	300	40
M1279	3.393 X 10^{10}	3.227 X 10^{13}	7795	77	40
M1280	4.750 X 10^{11}	2.829 X 10^{14}	373	300	15
M1280	2.859 X 10^{11}	1.790 X 10^{14}	8272	77	15
M1281	3.123 X 10^{9}	3.123 X 10^{13}	640.8	300	20
M1281	6.962 X 10^{9}	6.962 X 10^{13}	864.3	77	20
	note : n-type - 550 °C				

Bragg Mirror 20-µm GaAs w/ $Al_{.5}Ga_{.5}As$ Barriers

Sample #	Sheet Carrier Conc (cm^{-2})	Carrier Conc (cm^{-3})	Mobility (cm^2/ V-sec)	Temp (K)	As/Ga ratio
M1284	5.730 X 10^{11}	3.360 X 10^{14}	358	300	20
M1284	1.594 X 10^{11}	1.077 X 10^{14}	7843	77	20

Samples M1278, M1279, M1280, and M1284, that were grown at 595 °C, are p-type with a carrier concentration around 1–3 × 10^{14} cm^{-3}. In contrast, the sample M1281, which was grown at 550 °C, is n-type and has the smallest mobility at 77 K. The 300 K and 77 K mobilities generally increase as the As/Ga flux ratio decreases. The sample with the BR has a smaller 77 K and 300 K mobility than the sample grown without the BR.

3. Analysis and Conclusion

Note that the estimated initial photoexcited carrier density at an excitation power of 6.5 mW is 1.3 × 10^{15} cm^{-3} and the background hole concentration in the GaAs layers is around 10^{14} cm^{-3}.

Both the 6.5 mW and the 1.79 mW PL excitation pulses result in a strong injection of carriers. All of the samples exhibit PL decay that is bimolecular and nonexponential (*6, 7*) over a period of around 40 ns after the excitation pulse. Due to the initial charge separation and screening of the electric field in the depletion region by the photoexcited carriers, fast surface recombination dominates the PL decay until the photoexcited carrier density decreases to a level where low-injection conditions apply and the PL decay becomes exponential (*8*). After the initial 40 ns, a single exponential PL decay is observed for these 20-µm GaAs DH structures, indicating that the photoexcited structure are in the low-injection regime. Taking into account self-absorption of photons (photon recycling) in the 20-µm GaAs layer, the PL decay time (the effective minority carrier lifetime) τ is given by (*3*)

$$\frac{1}{\tau} = \frac{1}{\tau_{nr}} + \frac{2S}{d} \tag{1}$$

where S is the GaAs/AlGaAs interface recombination velocity and d = 20 µm is the thickness of the GaAs layer. The above equation shows that the effective minority carrier lifetime of the DH structures is primarily determined by the bulk nonradiative recombination time for typical values of S. Using the previously determined value for the interface recombination velocity S = 75 cm/s at the GaAs/AlGaAs interface (see ARL-TR-6186 [*2*]), we determine τ_{nr} from the observed PL decay time. Self-absorption of emitted photons in the 20-µm GaAs layers results in a small PL signal-to-noise ratio that precludes the use of a low enough excitation power, which would result in a low injection excitation regime and eliminate the fast surface recombination effects. The lifetime data for the samples is shown in table 2.

Table 2. Lifetime data.

Sample No., MBE Parameters	PL Decay Time	Nonradiative Lifetime, τ_{nr}
M1278, As/Ga = 20, 595 °C	871 ns	932 ns
M1279, As/Ga = 40, 595 °C	576 ns	602 ns
M1280, As/Ga = 15, 595 °C	1.12 µs	1.2 µs
M1281, As/Ga = 20, 550 °C	250 ns	255 ns
M1284, As/Ga = 20, 595 °C, Bragg Reflector	919 ns	987 ns

These results show that the optimum growth temperature is around 595 °C and that the nonradiative lifetime increases as the As/Ga flux ratio is lowered but the growth does not become arsenic deficient. The results confirm that the deep level trap densities in GaAs grown by MBE at 595 °C decrease as the As/Ga flux ratio is lowered. The longest τ_{nr} is obtained at the growth temperature of 595 °C with the As/Ga flux ratio = 15. The DH structure with the BR has a 6% increase in τ_{nr} indicating that growth of the BR results in a slight decrease in the deep level trap density in the 20-µm GaAs layer. Further studies are needed to confirm this. Previous research on a GaAs/AlGaAs DH structure with a 16-µm GaAs layer grown by liquid-phase-epitaxy showed that the lifetime ≈1.3 µs[3]. GaAs/ AlGaAs DH structures with 2–10-µm GaAs layers grown by MBE and organometallic chemical vapor deposition exhibited lifetimes over the range 0.25 µs–0.5 µs and 1 µs–2 µs, respectively (*7*). Our results show that sample M1280's

lifetime is comparable to the best reported lifetimes in MBE-grown undoped GaAs. The p-GaAs/AlGaAs DH structure with hole concentration ≈3 × 10^{16} cm^{-3} that was grown at ARL at 550 °C with the As/Ga flux ratio = 40, has a measured internal radiative quantum efficiency = 0.68. In contrast, by growing the same structure at ARL at 595 °C with the As/Ga flux ratio = 20, the internal radiative quantum efficiency increased to 0.9. We plan to investigate the possibility of a further increase in the internal radiative quantum efficiency by growing the GaAs/AlGaAs DH structure at a lower As/Ga flux ratio. The estimated error in the PL decay time is around 10% due to the TRPL measurement system characteristics, so further measurements may be carried out to confirm these results.

In conclusion, we report a significant increase in the nonradiative lifetime and the internal radiative quantum efficiency of a MBE-grown GaAs/AlGaAs DH structure that was grown at ARL using a substrate growth temperature of 595 °C and the As/Ga flux ratio = 15. Our results show that the nonradiative lifetime and the internal radiative quantum efficiency of the DH structures grown using these parameters are comparable to those of the highest quality reported MBE-grown GaAs.

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List of Symbols, Abbreviations, and Acronyms

AlGaAs	aluminum/gallium arsenide
ARL	US Army Research Laboratory
As	arsenic
BR	Bragg reflector
DH	double heterostructure
Ga	gallium
GaAs	gallium arsenide
MBE	molecular beam epitaxy
PL	photoluminescence
S	surface recombination velocity
Si	silicon
TRPL	time-resolved photoluminescence
τ	effective minority carrier lifetime
τ_{nr}	nonradiative lifetime
τ_r	radiative lifetime

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